

# **Enriched Oxygen Coal-Fired Combustion**

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## **Introduction**

Worldwide, coal consumption for electric power generation represented about 25% of the electricity production in 1996, while for USA and Canada it represented 57% and 19% respectively. In the case of Canada the coal used for electricity production varies considerably in the different provinces. If one also considers its large reserve compared to the other fossil fuels, coal will certainly continue to play a major role in energy production in North America and worldwide. In the past decade there has been a growing concern about greenhouse gas emissions ( $\text{CO}_2$ ,  $\text{CH}_4$ ,  $\text{N}_2\text{O}$ ) and its potential impact on climate change. Since coal-fired power plants account for about 11% of  $\text{CO}_2$  emission worldwide, it has a significant impact on the global greenhouse gas effect. To reduce  $\text{CO}_2$  emissions, it is necessary to develop clean and efficient combustion technologies for existing and new coal-fired power generation plants.

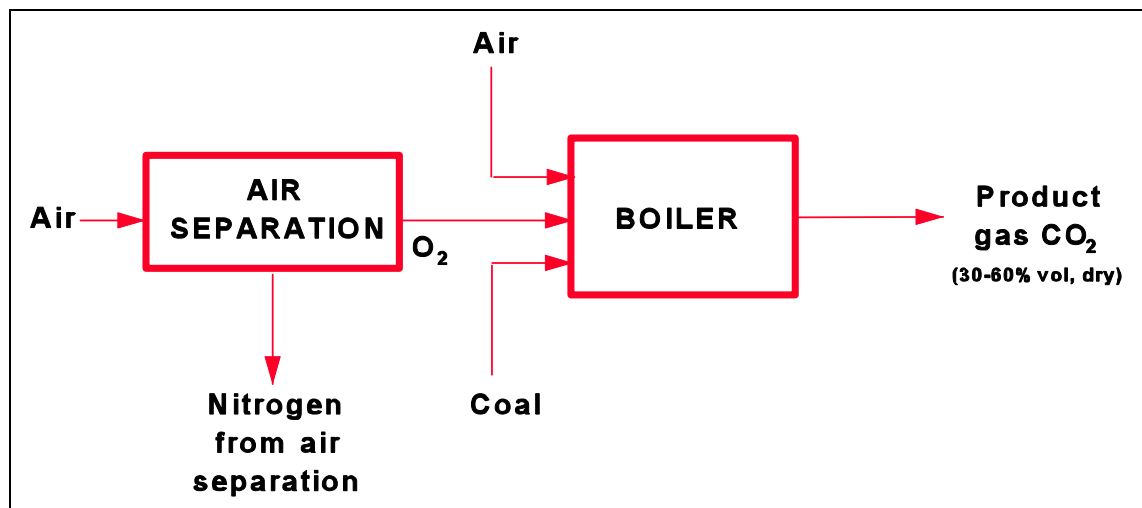
The most radical measure to reduce  $\text{CO}_2$  is to capture and store it. As indicated in Table 1, the storage capacity of  $\text{CO}_2$  in sinks worldwide and of a province such as Alberta in Canada is considerable. Table 1 also points out the potential market for  $\text{CO}_2$  use in enhanced oil recovery (EOR) and enhanced coal bed methane (CBM). The use of  $\text{CO}_2$  for EOR has now become a commercial reality in Canada. PanCanadian Petroleum of Calgary buys  $\text{CO}_2$  from the Great Plains Synfuel Plant in North Dakota for use in its oilfield in Weyburn, Saskatchewan. Unfortunately, capturing  $\text{CO}_2$  from conventional pulverized coal-fired power plant is currently very costly, in part because of its dilution in the flue gas and also because of the required pre-treatment of the flue gas. One option to reduce the cost of gas separation is to increase the concentration of  $\text{CO}_2$  in the flue gas. This can be achieved by increasing the oxygen in the feed gas and, eventually, by recirculating part of the flue gas. Coal combustion at higher oxygen concentration is particularly attractive because, not only does it allow reducing the cost of  $\text{CO}_2$  separation in the flue gas, but also because it reduces the volume of inert gas, such as nitrogen, and thus increases the boiler thermal efficiency. Recirculation of the flue gas is needed when a very high  $\text{CO}_2$  concentration in the flue gas is required.

**Table 1** - Worldwide and Alberta CO<sub>2</sub> storage capacities. <sup>a</sup>(y) estimated years of storage of national emissions based on Canadian CO<sub>2</sub> emissions at 1990 levels. Canadian 1990 emissions: 0.16 GtC. Global 1990 emissions: 7.5 GtC.

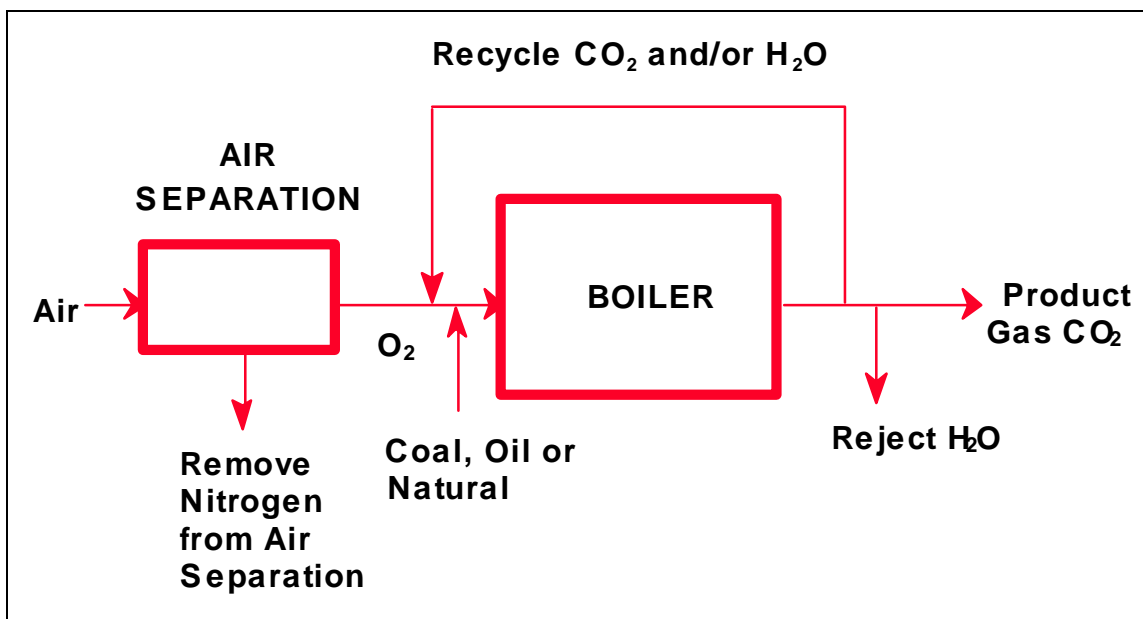
	Alberta <sup>a</sup> Gt of C	Global Gt of C
Ocean	?	> 1000
Aquifers	5.5 (35 y)	> 100
Depleted oil & gas fields	1 (6 y)	> 200
Enhanced oil recovery (EOR)	0.28 (2 y)	65
Coal bed methane (CBM)	2.7 (17 y)	100 ?

### Application

Potential power generation schemes employing enriched oxygen with or without flue gas recirculation are shown in Figures 1 and 2. Figure 1 represents a schematic for enriched air coal-fired combustion without flue gas recirculation. The principle is to mix the secondary air with pure oxygen. Depending on the level of oxygen enrichment, the concentration of CO<sub>2</sub> in the flue gas can reach between 55 and 60% vol., dry. Such high concentrations of CO<sub>2</sub> in the flue gas may be enough for less costly separation of CO<sub>2</sub> in post combustion treatment operations or for disposal in geological media.



**Figure 1** - Enriched air coal-fired combustion scheme



**Figure 2** - Flue gas recycle combustion applied to a steam boiler

Figure 2 shows a schematic for enriched oxygen coal-fired combustion with flue gas recirculation. The application shown in this figure is intended for use in a new or retrofit modification to a steam boiler burning not only pulverized coal, but also oil or natural gas. In such a power generation application, the resulting effluent is a CO<sub>2</sub>-rich flue gas (up to 98% vol., dry) that can be removed for utilization or disposal with minimum additional flue gas treatment.

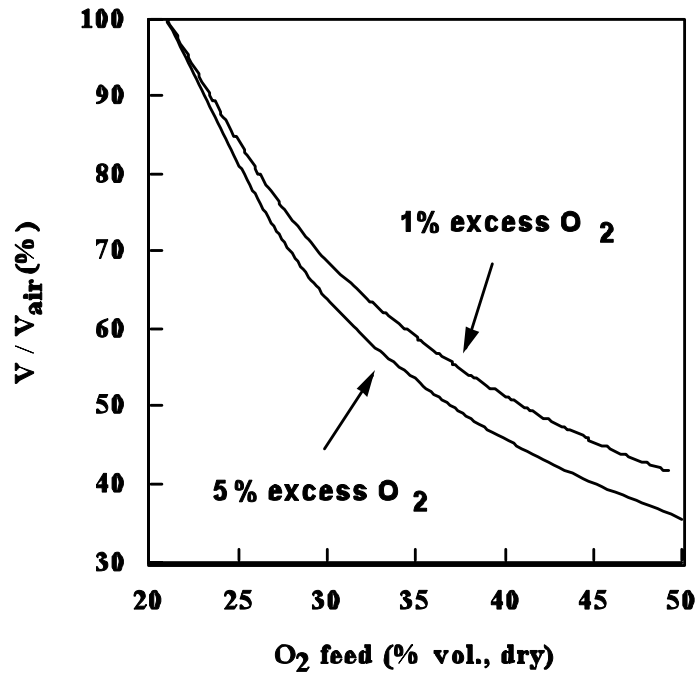
Any of the direct coal-fired schemes described above could be built as a demonstration plant in North America. Depending on the nature of future regulations to reduce or cap greenhouse gas emissions from industrial or utility power plants, the technology can be applied to produce a concentrated stream of CO<sub>2</sub> that could be directly removed for utilization or storage in land based sites. However, the initial application of the technology will very likely be in its use to retrofit or re-power existing pulverized coal-fired power stations. In that regard, it is likely that the enriched oxygen coal-fired combustion scheme without flue gas recirculation will initially attract more attention for retrofitting existing power plants because it should involve much less modification than the flue gas recirculation scheme. However, in the enriched oxygen coal-fired combustion scheme the CO<sub>2</sub> concentration in the flue gas depends directly on the oxygen concentration in the feed gas; the higher the O<sub>2</sub>

concentration in the feed gas, the higher the  $\text{CO}_2$  concentration in the flue gas. It is anticipated that higher oxygen concentration in the feed gas will lead to higher flame temperature, which might be well above the ash fusion temperature, causing potential slagging problems. Other problems posed by the high temperature on existing boiler materials can also be anticipated. The point is that the level of oxygen in the feed gas will very likely be limited. The maximum oxygen concentration allowed will set the maximum  $\text{CO}_2$  concentration in the flue gas. It is now premature to give an estimate of the maximum oxygen concentration in the feed gas for applications in an existing or new coal-fired boiler, and more research is needed in that direction. If the focus is on high  $\text{CO}_2$  concentration in the flue gas, the oxygen level in the feed gas is less an issue in the case of coal combustion with flue gas recirculation. Indeed, very high  $\text{CO}_2$  concentration in the flue gas (up to 98% vol., dry) can be achieved, regardless the oxygen concentration in the flue gas. In that respect enriched air coal-fired combustion with some flue gas recirculation can also be of interest from the perspective of retrofitting existing power plants.

There are several benefits for  $\text{O}_2/\text{CO}_2$  recycle combustion. First of all, concentration up to 98%  $\text{CO}_2$  in dry flue gas may be attainable, allowing  $\text{CO}_2$  to be reused or stored with no or little extra treatment. The purity of the  $\text{CO}_2$  stream will be imposed by the downstream  $\text{CO}_2$  application. For example,  $\text{CO}_2$  storage in aquifers will likely require less stringent purity criteria than the use of  $\text{CO}_2$  for EOR or CBM. The  $\text{CO}_2$  purity requirement is also closely related to oxygen purity after air separation. Generally, a cheaper and lower purity oxygen separation process will produce a lower purity  $\text{CO}_2$  flue gas stream from this modified combustion process. If this route is chosen and a higher  $\text{CO}_2$  purity flue gas stream is desired, more costs will be incurred from the need to further process the flue gas.

Another important benefit is the reduced volume involved due to combustion at higher oxygen concentration, and thus with less inert gas volume (mainly  $\text{CO}_2$  at the expense of reducing  $\text{N}_2$  in the present case). The advantages of this reduced volume are lower dry gas energy losses, higher plant efficiency and lower energy loss for gas cleaning/separation. Figure 3 shows the ratio of the required volume of gas in the case of combustion with recycled flue gas over the required volume in the case of combustion in air. This ratio is shown for different concentrations of oxygen in the feed gas. Figure 3 is for Eastern bituminous coal burning at constant thermal input for the two cases of 1% and 5% excess oxygen in the flue gas. Note that this figure is for combustion with flue gas recirculation, but it is very similar in the case of enriched air combustion. Combustion in air and in 21%  $\text{O}_2$  with flue gas recirculation are equivalent in terms of volume of gas required (ratio of 100% in the graph). However, an increase in the oxygen feed decreases significantly the required volume of gas. For example, the volume of gas required for combustion at 40% oxygen represents only between 45 and 50% of the required volume for combustion in air. For comparison, Figure 3 shows the cases of 1% and 5% excess oxygen. This figure shows that the 5% excess oxygen situation requires lower volume of gas than for the 1% excess oxygen case. In fact the difference between these two cases is in the volume of pure oxygen brought into the system. For the 5% excess oxygen case, less volume of pure oxygen is needed because more oxygen is brought back to the system through recirculation of the flue gas than in the case of 1% excess oxygen. Another advantage associated with the higher oxygen concentration is the ability to minimize unburned carbon.

Another benefit of  $O_2/CO_2$  recycle combustion is the expected reduction of  $NO_x$  emission. Indeed, Japanese researchers<sup>1,2</sup> found that  $NO_x$  emissions are lower during  $O_2/CO_2$  combustion and recycled combustion. They explain  $NO_x$  reduction during recycled combustion by the fact that part of the recycled NO (actually more than 50%) is reduced to molecular nitrogen ( $N_2$ ).



**Figure 3** - Ratio of the required volume of gas in the case of combustion with recycled flue gas over the required volume in the case of combustion in air. Case of Eastern bituminous coal.

### Objective and Approach

Coal flame characteristics at high oxygen concentration and at high  $CO_2$  concentration, as in the case of flue gas recirculation, are not well understood. The objective of this work is to investigate enriched oxygen coal combustion when part of the flue gas is recirculated in order to maximize  $CO_2$  concentration in the flue gas prior to its recovery. More precisely, this work aims at studying the

influence of oxygen concentration in the feed gas and the effect of flue gas recirculation on key combustion parameters such as flame temperature and pollutant formation ( $\text{NO}_x$ , and  $\text{SO}_2$ ).

The approach chosen is to perform experimental and modeling work, the two interacting with each other. The experimental work is used initially to extract the main features of a coal flame at high  $\text{O}_2$  and  $\text{CO}_2$  concentration. These experiments are also used to validate a coal combustion model that is currently under development at CETC. Through the model it is intended to extrapolate the results obtained at a pilot scale to a power plant boiler size. In this paper only results concerning the experimental work will be presented.

The experimental study is done on a pilot-scale process designed to undertake work on  $\text{O}_2/\text{CO}_2$  recycle combustion at CANMET Energy Technology Centre in Ottawa, Canada. A cylindrical, down-fired vertical combustor was chosen as the preferred method of fuel combustion. This reactor permits the establishment of axisymmetric flames that can be analyzed by water-cooled instruments and probes to evaluate the radial and axial profiles of in-flame gaseous and particulate species, temperature, and heat transfer properties. The vertical combustor has a rated firing capacity of up to 0.3 MW and can burn solid, gaseous and liquid fuels. However, to the present time only coal and natural gas have been tested in the reactor. Figure 4 provides a simplified schematic of the vertical combustor facility. The combustor is made up of cylindrical lined sections with a 1.0 m outer diameter and a 0.60 m inner diameter. The whole assembly is made up of 11 sectional pieces with an overall length of 6.7 m.

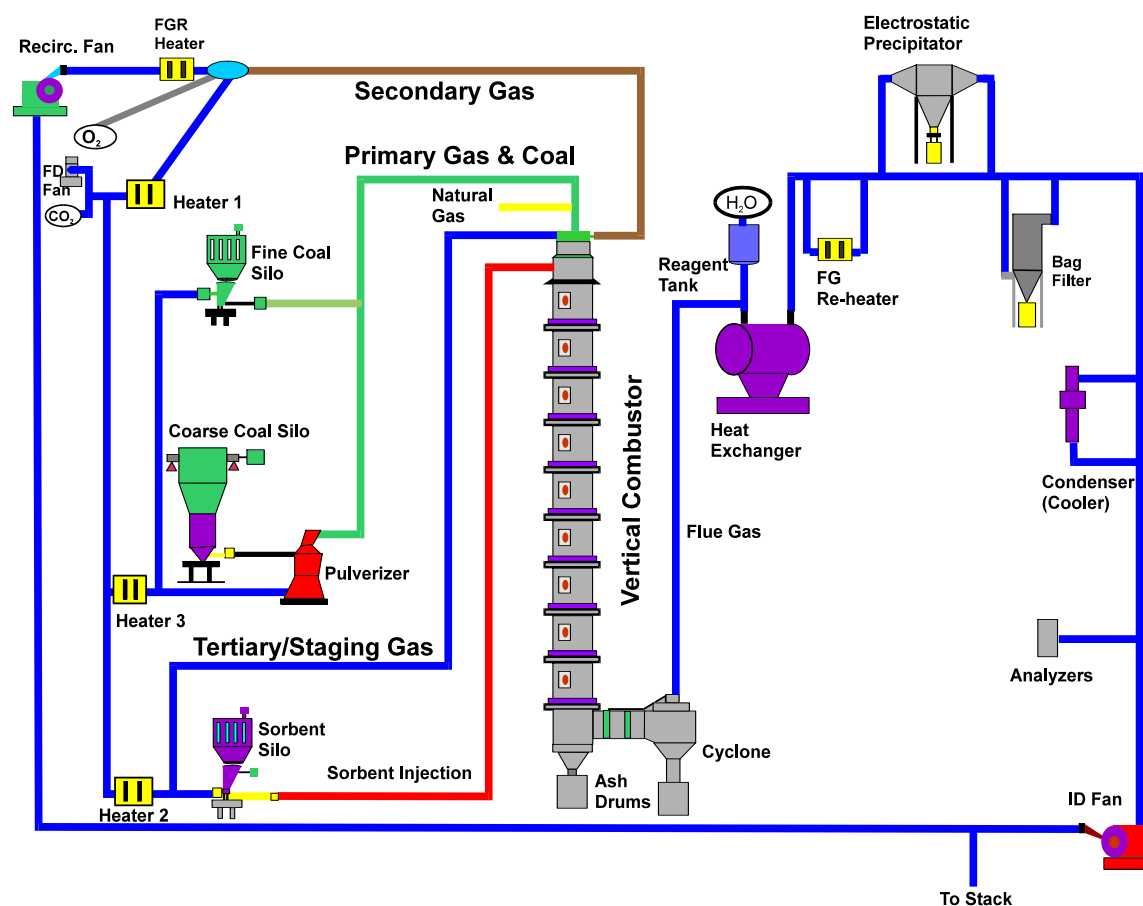
The upstream gas distribution system to the combustor provides air flow from a forced draft fan for plant operation in the air-blown combustion mode. Mixture of carbon dioxide and oxygen to simulate  $\text{O}_2/\text{CO}_2$  recycle combustion can also be supplied as the combustion medium. In this instance,  $\text{CO}_2$  and  $\text{O}_2$  are obtained from 50 ton cryogenic liquid storage tanks located within the laboratory complex. The upstream piping setup also provides for the use of a third type of combustion medium using recycled flue gas mixed with oxygen.

Coarse coal, dried and crushed to minus 30 mm, is stored in a hopper mounted on a weigh scale and fed to a ring-roller type of pulverizer. The pulverizer is capable of operating at throughputs of up to 80 kg/h of coal. A variable speed air classifier located on the pulverizer outlet controls the fineness of the pulverized coal size grind. Pulverized coal crushed to the desired fineness specifications is pneumatically conveyed to the coal burner when direct firing the coal, or is diverted to a fine coal hopper for storage and re-injection when indirectly firing the coal to the combustor. In the indirect firing mode, fine coal stored in a weigh hopper under  $\text{CO}_2$  purge is re-fed to the burner via a variable speed, twin screw solids feeder and a venturi eductor.

In the downstream section of the plant, the flue gases leaving the combustor pass through a refractory lined cyclone for the removal of coarse particulates. The gases undergo cooling in a glycol-filled boiler that serves as a heat exchanger. The cooling can be adjusted by changing the number of passes through the heat exchanger.

Once cooled, the gases pass through a flue gas plenum from which they can be routed through a number of different types of gas cleaning devices. The first device is an electrostatic precipitator (ESP) used for fine particulate removal. The second device is a bag or fabric filter unit for fine particulate control. The unit relies on the use of compressed air or CO<sub>2</sub> for on-line reverse pulse-jet cleaning of individual bags. The ash filter cakes removed during cleaning are collected in an ash hopper prior to discharge from the bag house unit. A flue gas desulphurisation (FGD) unit can also be connected prior to the ESP. In conjunction with upstream flue gas humidification in the boiler, the FGD unit can be operated either with conventional lime sorbents or narcholite (sodium bicarbonate) injection for SO<sub>2</sub> capture.

During plant operation, the exit flue gas is monitored to determine the composition of CO, CO<sub>2</sub>, NO<sub>x</sub>, SO<sub>2</sub> and O<sub>2</sub> using on-line gas analyzers. The pilot plant ambient is sampled continuously to determine the oxygen concentration, mainly to monitor any CO<sub>2</sub> leakage and to avoid any safety hazards for the plant personnel. When ambient leakage of CO<sub>2</sub> is low, the combustor and the downstream flue gas plenum is operated under positive pressure so as to minimize ambient nitrogen contaminant of the process gas.



**Figure 4** – Schematic of the pilot-scale facility (“Vertical Combustor”)

Extensive plant supervisory and data acquisition for the vertical combustor facility is performed by a Honeywell Series S9000e controller. This controller, along with its dedicated PC-based monitor, allow plant personnel to run the fuel and gas delivery system to the combustor from a control room in the pilot-plant while also monitoring all process measurements.

## Results

All experiments were performed at a firing rate of 0.2 MW (0.7 MBtu/h) for Eastern bituminous coal. The proximate and ultimate analysis of the coal is given in Table 2. We can distinguish three types of runs: combustion in air, combustion in an  $O_2/CO_2$  mixture and combustion with flue gas recirculation. Combustion in air represents the reference case. For the experiments in  $O_2/CO_2$  mixtures, the nitrogen is replaced by  $CO_2$ . The main purpose of this type of experiments is to understand the effect of high  $CO_2$  concentration, and also high  $H_2O$  concentration on flame characteristics. The experiments with flue gas recirculation are done to study the effect of recycling the flue gas and to mimic the conditions of future applications. Every experiment is done at 5% excess oxygen, that is 5% vol., dry of oxygen in the flue gas. The oxygen in the feed gas varies between 28 and 42% vol., on a dry basis. Also, to simulate the effectiveness of the air separation unit, experiments are done for an oxygen purity of 100% (ideal case) and 90%. An oxygen purity of 100% means that a pure stream of oxygen comes out of the air separation unit, whereas an oxygen purity of 90% means that this stream contains 90%, on a dry volume basis of oxygen, the remaining 10% being nitrogen. The distinction between these two cases is important because the presence or absence of nitrogen affects the  $NO_x$  chemistry and also affects the separation process of  $CO_2$  in the flue gas. Note that in the present work, there is no separation unit. Mixing the right quantity of pure oxygen and pure nitrogen simulates the effectiveness of the air separation.

**Table 2.** Proximate and ultimate analysis of Eastern bituminous coal

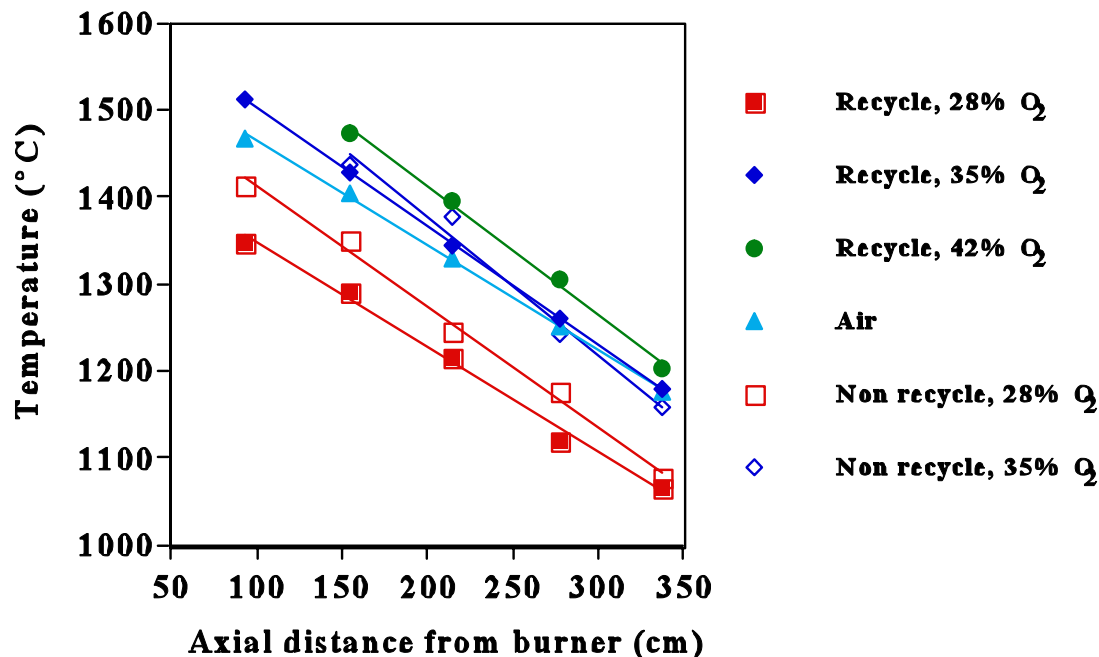
	Eastern bituminous coal
Moisture, as received (wt%)	1.58
<b>Proximate analysis (wt %, dry)</b>	
Ash	8.41
Volatile	35.24
Fixed carbon (by diff.)	56.35
<b>Ultimate analysis (wt %, dry)</b>	
Carbon	77.65
Hydrogen	5.01
Nitrogen	1.45
Sulfur	0.96
Ash	8.41
Oxygen (by difference)	6.52
Heating value (MJ/kg, dry)	32.2



In the case of experiments with flue gas recirculation, two cases are possible: dry and wet experiments. In the case of dry experiments, the water in the flue gas is condensed and removed before being recirculated. In the case of wet experiments all the water in the flue gas is recirculated. The distinction between dry and wet experiment will allow us to observe the influence of water on the flame characteristics. For the wet configuration, the concentration of water can go up to 18% vol. For comparison, combustion in air for similar conditions of excess oxygen and oxygen in the feed gas leads to about 5% water in the flue gas. The analysis from the wet experiments are not all completed at the current time. Therefore, only results concerning dry experiments are shown in the present paper.

## Flame temperature

Figure 5 shows the suction pyrometer measurements of the centreline flame temperatures in the vertical combustor. This figure indicates that, whatever the type of enriched oxygen experiment (recycle or not), the flame temperature increases when the oxygen concentration increases in the feed gas. If one looks at a distance of 150 cm from the burner, the temperature rises from about 1300°C at 28% O<sub>2</sub> to about 1500°C at 42% O<sub>2</sub>. However, combustion in air (20.9% O<sub>2</sub>) leads to higher flame temperature than combustion at 28% O<sub>2</sub>. As noted by Miyamae et al. (1994), the higher specific heat of the CO<sub>2</sub> gas compared with nitrogen will lead lower flame temperatures than for an equivalent oxygen concentration in an air combustion medium.

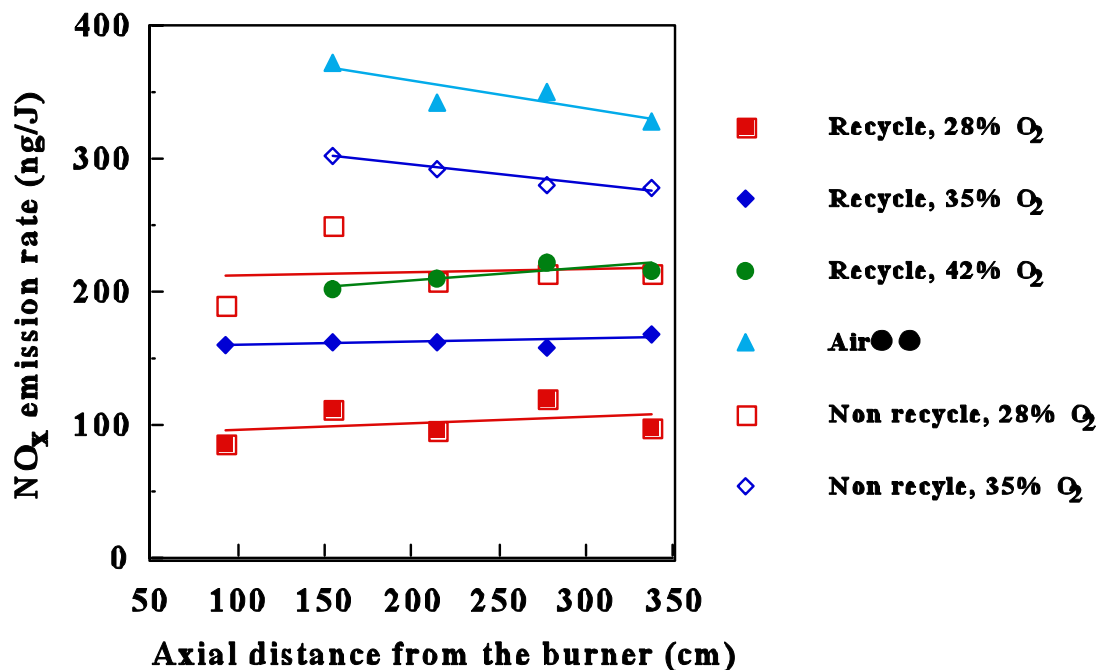


**Figure 5** - Centerline axial temperature. Non recycle represents experiments in O<sub>2</sub>/CO<sub>2</sub> mixtures. Recycle runs are only dry experiments.

For 35% O<sub>2</sub> there is no difference between non recycle experiments and dry recycle one, which might be expected because all experimental conditions are actually very close in both cases. The difference observed at 28% O<sub>2</sub> needs to be further investigated. Figure 5 also shows that the oxygen concentration, for either recycle or non recycle tests, generating the same flame temperature as for combustion in air lies between 28% and 35% vol. of O<sub>2</sub> in the feed gas. These values are somewhat lower than that of about 42% given by Miyamae et al.<sup>1</sup> based on their calculations of the adiabatic flame temperature.

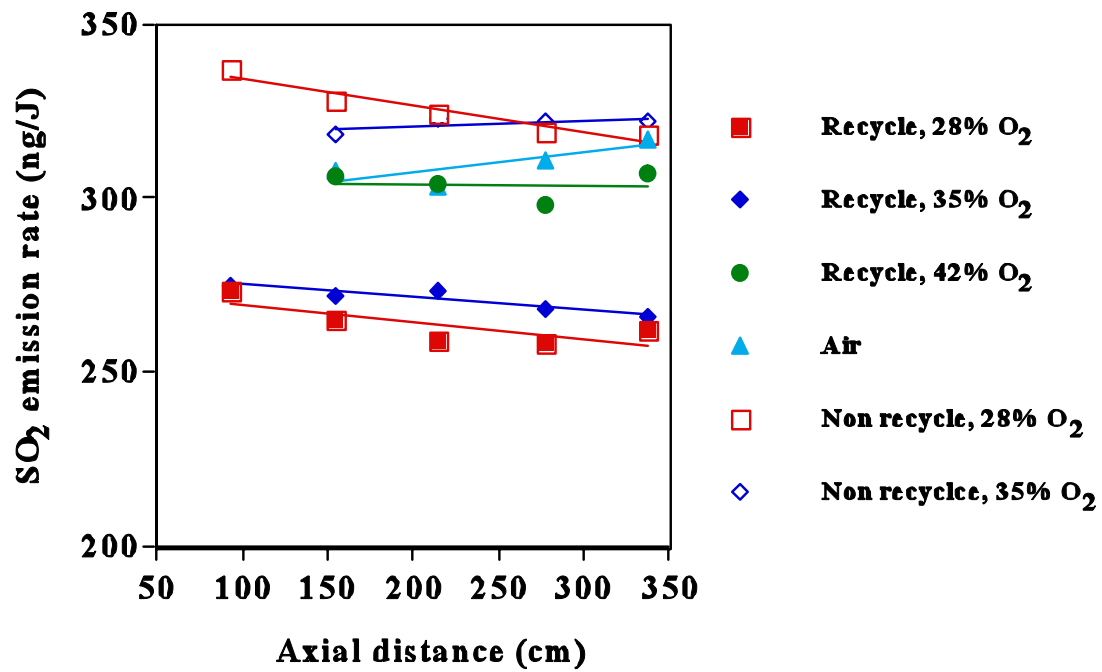
### **NO<sub>x</sub> and SO<sub>2</sub> emissions**

Data for NO<sub>x</sub> and SO<sub>2</sub> emissions are expressed as a pollutant emission rate (ng/J) and represent the mass of pollutant emitted per unit heat produced by the coal. Figure 6 shows the centerline axial NO<sub>x</sub> emission rate. This figure indicates that higher oxygen concentration in the feed gas increases the NO<sub>x</sub> emission rate. In fact, the influence of oxygen has to be linked to the influence of temperature. In the previous section it was mentioned that an increase in O<sub>2</sub> feed leads to higher flame temperature which, as a result, increases the NO<sub>x</sub> emission rate. Indeed, higher temperatures favor both homogeneous and heterogeneous NO<sub>x</sub> formation. Another important observation is that the highest NO<sub>x</sub> emission rate was found in the case of combustion in air. This can be explained by higher formation of thermal NO<sub>x</sub> due to more molecular nitrogen in the system than in the case of recycle and O<sub>2</sub>/CO<sub>2</sub> combustion mixtures. Figure 6 also shows that combustion with flue gas recirculation considerably reduces the NO<sub>x</sub> emission rate compared to combustion in O<sub>2</sub>/CO<sub>2</sub> mixtures. This finding is in agreement with the observation of Miyamae et al.<sup>1</sup> and Okazaki et al.<sup>2</sup> who explained NO<sub>x</sub> reduction during recycled combustion by the fact that part of the recycled NO is reduced to molecular nitrogen (N<sub>2</sub>). Another factor that can enhance NO<sub>x</sub> reduction is its dissolving in the condensed water, and hence removal from the gas stream, during dry recycle tests.



**Figure 6** – Centreline axial NO<sub>x</sub> emission rate.

Figure 7 shows the centreline axial SO<sub>2</sub> emission rate. In the case of O<sub>2</sub>/CO<sub>2</sub> mixture runs SO<sub>2</sub> emission rate is not affected by the oxygen concentration in the feed gas. The value of the SO<sub>2</sub> emission rate in the case of combustion in air is very similar to the one found in combustion in O<sub>2</sub>/CO<sub>2</sub> mixtures. This result suggests that SO<sub>2</sub> chemistry is not affected by the presence of high concentration of CO<sub>2</sub> and O<sub>2</sub>, at least in the range 28 – 35% for the oxygen. At 28 and 35%, dry recycle experiments show much lower SO<sub>2</sub> emission rates than for non-recycle runs. Because we have seen that in this concentration range, CO<sub>2</sub> and O<sub>2</sub> has no particular effect on SO<sub>2</sub> formation, it is suspected that the main difference in SO<sub>2</sub> emission rate between recycle and non recycling tests is due to SO<sub>2</sub> dissolving in the condensed water and thus removed from the system. This process has already been mentioned to explain part of the NO<sub>x</sub> reduction. However, compared to the NO<sub>x</sub> case, where it is probably minor compared to homogeneous NO<sub>x</sub> reduction, this dissolving process is probably the main factor for SO<sub>2</sub> reduction. This is supported by the fact that a concentration of more than 1000 ppmv of sulfate has been found in the condensed water.



**Figure 7** – Centerline axial SO<sub>2</sub> emission rate.

## **Future Activities**

Present and future activities for this work are listed as follows:

- Enriched air combustion tests
- Recycle combustion tests at lower excess oxygen
- Numerical simulation of the reactor (CFD modeling)
- Integrated process modeling for enriched air combustion plant and flue gas recirculation plant
- Design, installation and test of a condensor unit for flue gas separation
- Material corrosion and slagging and fouling tests

## **Acknowledgement**

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2. Okazaki, K. and Ando, T., NO<sub>x</sub> Reduction Mechanism in Coal Combustion with Recycled CO<sub>2</sub>, Energy Vol. 22, No. 2/3, pp. 207-215, 1997.